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Modeling of transport phenomena in battery cathode materials

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Transport phenomena in the cathode materials of batteries, both ionic and electronic, play a key role in the overall performance of the device. An intense modeling activity of different aspects of these transport phenomena has taken place during the last two decades, which covers different time and length scales. In this talk we will focus on modeling at the atomic scale of ionic and electronic transport in the cathode materials of lithium-ion batteries and aprotic metal-air batteries.

The ionic transport of intercalated lithium in different transition metal oxides has been extensively modeled in the literature by combining density functional theory (DFT) and the nudged elastic band (NEB) method to obtain the activation barriers for lithium diffusion within these materials.[1] Lately the method is starting to be applied also to the ionic transport between coating materials (e.g. graphite) and the cathode and also at the interfaces that are created between different phases of the same cathode material[2]. During the talk we will present some examples of lithium transport through these interfaces.

The electronic transport in lithium-ion batteries typically occurs through a polaronic hopping mechanism, which is conventionally modeled using DFT and NEB as in the case of the ionic transport. This way of modeling the electronic transport is valid in the purely adiabatic limit, omitting all the non-adiabatic features of the charge transfer. Here we will briefly introduce our recent developments in the implementation of a DFT-based Marcus theory model which is able to capture the non-adiabatic effects.

In contrast with lithium-ion batteries a conversion process takes place in metal-air batteries. In this conversion process an oxide is formed at the cathode through the reaction between atmospheric oxygen and alkali or alkali-earth ions. We will talk about modelling the formation of these oxides and its relation with the overpotentials observed in these batteries[3].

Finally we will address the electronic transport at metal-air battery cathodes, which can take place through polaronic hopping[4] as in the case of lithium-ion batteries, but also can occur through electron tunneling[5]. We will discuss which of the two processes is dominant depending on the different conditions of the charge/discharge process.

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